

Microstructure and properties of polymeric composite materials based on polyethylene and thermally extended graphite for transport systems

E.A. Lysenkov, L.P. Klymenko*

*Petro Mohyla Black Sea National University;
10, 68 Desantnykiv Str., Mykolaiv, 54003, Ukraine*

Received: 30.05.2021 Accepted: 06.06.2021

Abstract

The effect of thermoexpanded graphite (TEG) on the microstructure and functional properties of polymer composites based on high-density polyethylene (HDPE) has been studied by optical microscopy, differential scanning calorimetry and mechanical analysis. There has been proven the efficiency of the method of mixing polymer composites using a piston extruder, which provides a more uniform distribution of the filler in the polymer matrix. It is shown that the introduction of TEG leads to a decrease in the degree of crystallinity and melting point of systems based on high-density polyethylene, which is a consequence of the destruction or increase in the defect of the crystal structure of the polymer matrix under the influence of TEG. With the introduction of 1 % of TEG, the melting point decreased from 415.0 to 408.5 K. With the introduction of 3 % of TEG, the thermal conductivity increased from 0.18 W/(m·K) (for HDPE) to 0.76 W/(m·K). The extreme change in the thermal conductivity of polymer composites is a consequence of the formation of TEG in the polymer matrix of the percolation cluster, the mesh of the filler, which penetrates the entire volume of the material. As a result of the conductivity studies, the percolation threshold of thermal conductivity has been determined for these HDPE-TEG systems, which is 0.6 %. Microscopic studies confirmed the formation of the percolation cluster obtained by thermal conductivity. It is shown that at the content of 0.6 % of TEG, there is formed a continuous cluster. The formation of this cluster is confirmed by mechanical studies. An increase in mechanical strength has been recorded, which increases from 30.5 MPa (for HDPE) to 42.8 MPa at 5 % filler content, and this is promising for the use of these materials in transport systems.

Keywords: *high-density polyethylene, percolation, polymer composite material, thermoexpanded graphite.*

Introduction

In recent years, the development of new polymer composites has attracted considerable attention due to their potential application in advanced technologies, for example, in antistatic, corrosion-resistant coatings, transportation systems, sensor microelectronics, and other areas of production [1]. The introduction of fillers with unique properties, such as graphite, carbon black, powders of metals and metal oxides of various types and sizes into a polymer matrix, is a promising approach for the manufacture of polymer materials with improved functional characteristics. Such composites can have enhanced physical characteristics such as increased rigidity, strength, barrier properties, and heat resistance, without loss of impact strength over a very wide range of conventional synthetic or natural polymers [2].

Thermally expanded graphite (TEG) is one of the promising fillers for making such composite systems. Polymer-based composites, reinforced with expanded graphite [3], demonstrate significantly improved mechanical, electrical and other thermophysical

properties compared to unmodified polymer. The reason for this is the plate-like structure of natural graphite, where the atoms are firmly bound in the hexagonal plane, but weakly bound in the direction perpendicular to this plane. Therefore, when making thermally expanded graphite, these plates are separated, to a thickness of the order of several hundred nanometers, while they are characterized by a very large surface (up to 2630 m²/g). That is why the separation of TEG into plates and their uniform distribution in the polymer matrix is a difficult task, but its solution will have significant prospects for creating polymer composite materials with desired properties.

So, polymer materials containing thermally expanded graphite have significant application prospects for transportation systems, for example, pipelines. However, the creation and research of such materials is associated with a number of problems, one of which is the uniform distribution of filler particles in the polymer matrix. The solution to this problem for thermally expanded graphite makes it extremely promising filler and enables to create materials with improved functional characteristics.

For example, in work [4] there was used a HDPE and TEG system to create shape-stable materials with a high storage capacity of thermal energy and thermal conductivity. As a result of the study, it was shown that with a uniform distribution of TEG particles in the

* Corresponding author:
ealysenkov@ukr.net

HDPE matrix, the thermal conductivity of a material with 3 % of TEG increased to 0.67 W/(m·K), while the thermal conductivity of HDPE was only 0.19 W/(m·K). In addition, HDPE-TEG materials provide significant thermal energy storage capacity and high thermal conductivity for latent heat storage.

The studies, carried out in [5], have shown that composite materials based on polypropylene and 9 % of TEG have improved thermal conductivity, thermal stability and resistance to flammability of the material. It was also found that although TEG affects the mechanism of thermal destruction of the material under study, the graphite plates inhibited the release of volatile destruction products.

A significant increase in thermal conductivity was observed by the authors [6] upon the addition of thermally expanded graphite to HDPE. Thermal conductivity increased from 0.442 W/(m·K) for pure HDPE to 0.938 W/(m·K) for composites containing 7 % of TEG. The heat capacity increased with rising temperature for both pure HDPE and composites filled with thermally expanded graphite, and no noticeable difference in heat capacity values was found due to the particle size.

A significant improvement in properties can be obtained by introducing a very large amount of the filler. Thus, in [7], the authors made composites based on high-density polyethylene and thermally expanded graphite for heat exchangers. At a content of 50 wt. % of TEG, the thermal conductivity increased by 37.2 % compared to unfilled HDPE in the material. However, after 9 months, a significant deterioration in material properties was observed due to high defectiveness through microphase separation. It is also found in the work that when the content of TEG is below 30 %, it is advisable to use the extrusion method for manufacturing composites, while with the introduction of more than 30 %, the injection molding method is effective.

Thus, an analysis of the scientific literature has shown that studies of the effect of thermally expanded graphite on the properties of polymer materials are highly relevant. Therefore, the purpose of our research was to study the effect of thermally expanded graphite on the microstructure, thermophysical and mechanical properties of polymer composite materials based on high density polyethylene. Achievement of this goal involves the implementation of the following tasks:

1. Manufacturing of a number of polymer composite materials based on high density polyethylene and thermally expanded graphite by the extrusion method using a piston extruder.
2. Conducting microscopic researches to study the microstructure of the materials obtained.
3. Investigation of thermophysical characteristics, namely thermal conductivity and heat capacity, of HDPE-TEG systems and their analysis.
4. Carrying out mechanical tests for materials based on HDPE-TEG.

Objects and research methods

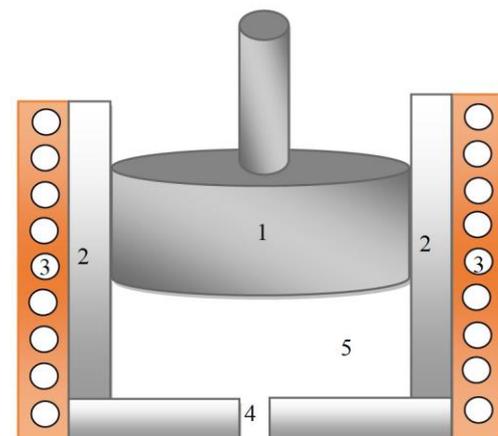
For the research, we used model systems based on high density polyethylene and thermally expanded graphite.

High-density polyethylene (HDPE), produced by SABIC®HDPE Eurotrubplast Holding Company Ltd Trusthose (Saudi Arabia), was chosen as a polymer matrix for the preparation of test samples ($T_{pl} \approx 415$ K).

For the manufacturing composite mixtures, thermally expanded graphite (TEG) powders with a bulk density of about 15 kg/m³ and a specific surface area of $\approx 2.5 \cdot 10^5$ m²/kg were used. In the structure of TEG, as shown by previous studies [9], there are nanosized extended formations with effective diameters (1–100) nm, which determines its high ability to form infinite clusters in polymer composite materials.

Composite samples were prepared by the extrusion method (mechanical grinding in the melt) using a piston extruder, followed by cooling under normal conditions. The content of the filler was varied in the range of (1÷5) wt. % (hereinafter – %). The main advantage of piston extruders over screw extruders is the ability to vary the mixing time of the filler particles with the molten polymer matrix, after which the test sample can be formed in the form of either a plate or a thread.

The manufacturing technology of materials studied is given below. After preliminary preparation of the composite material components by the method of mechanical mixing, the mixture was loaded into the extruder (Fig. 1). The mold was heated up to 200 °C. Rotation of the piston provides melt mixing of the polymer composite due to its own relatively high viscosity of the polymer.



1 – a piston that rotates; 2 – cylindrical base;
3 – heater; 4 – a hole in the base of a cylindrical shape;
5 – powdered composite

Figure 1 – Schematic representation of a piston extruder

It has been found experimentally that the most uniform mixing of filler particles in the matrix occurs within 10 minutes. With a further increase in the mixing time, there occurs partial degradation of the polymer. After 10 minutes, a hole is opened in the lower part of the mold, and without turning off the rotation of the

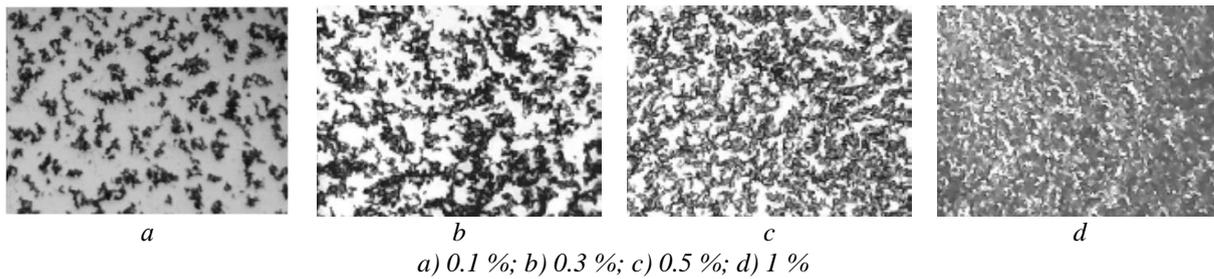


Figure 2 – Micrographs of HDPE-TEG systems. The magnification is $\times 40$

piston, the pressure of the translational motion gradually increases. The shear stresses that occurred caused the polymer composite melt to pass through the hole (~ 2 mm diameter). After that, the resulting material was placed in a hot-pressing mold in threadlike form, where the corresponding shape of the test sample was set.

The thermal conductivity of the samples under study was measured by the dynamic calorimetry method using an IT- λ -400 device (Kazakhstan) (thermal conductivity meter) with an improved cell [9]. The meter was calibrated by measuring the thermal conductivity of quartz and copper reference samples. The measurements were carried out in the monotonic heating mode. To improve the measurement accuracy, the thermal conductivity of each sample was measured three times with subsequent averaging of the results. The measurement error was 3 %.

Thermophysical studies were carried out in a dry air atmosphere using an upgraded PerkinElmer DSC-2 (Germany). The temperature range was from 300 K to 500 K, the heating rate was 2 K/min, and the research method was modulated by the DSC.

In this work, a study of tensile fracture stresses was carried out according to tensile strength. The dimensions of the sample studied were: length $l=100\pm 2$ mm, diameter $d=2\pm 0.3$ mm. The study of the effect of filler content on tensile strength was carried out using an automatic tensile testing machine UM-5 (Ukraine) (modified with high-precision pressure and displacement sensors) at a load speed $v = 5$ N/s. The measurement error was no more than 3 %.

Transmission micrographs of HDPE-TEG systems were obtained using an XY-B2 optical microscope equipped with an ICM 532 Digital Video Glasses and an AMCAP/VIDCAP image processing system (Microsoft). The samples under study were placed in a glass cell with a thickness of 80 μm .

Results and their discussion

Structural organization of TEG at the micro level.

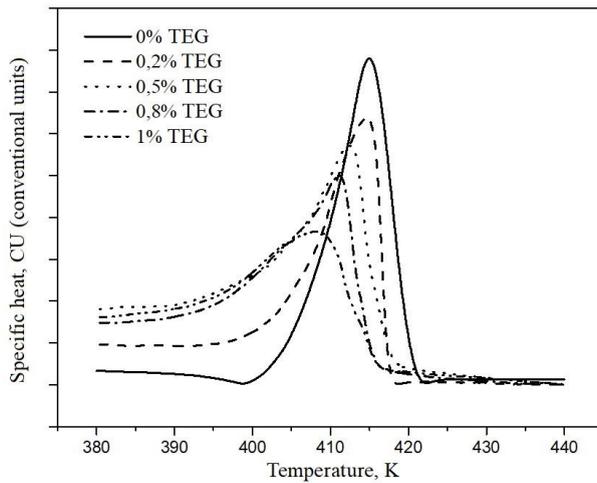
For the majority of polymer nanocomposites filled with TEG, with an increase in the filler content, the interactions between individual graphite plates become stronger than the polymer-TEG interaction. As a result, there is observed a typical percolation transition from a nonconducting state to a conducting one in such systems, at a certain threshold of TEG concentration. In this case, the TEG or their aggregates form a grid that permeates the entire volume of the studied material. Figure 2 presents microscopic images of HDPE-based

nanocomposites with various TEG concentrations in the range from 0.1 to 1 % at $T = 293$ K. Figure 2, *a-b* shows that when the content of TEG is close to the percolation threshold ($C = 0.1\text{--}0.5$ %), graphite plates form separate clusters that do not interconnect. If concentrations are near the percolation threshold ($C \approx 0.5$ %), TEGs begin to form large agglomerates. When the percolation concentration is reached, there is formed a continuous percolation cluster. At concentrations exceeding C ($C = 0.5\text{--}1$ %), TEG clusters begin to grow, forming more and more continuous channels (percolation network). Such a low value of the percolation threshold is typical for polymer-TEG systems and is explained by the high aspect ratio, $r = l/d$ ($r \approx 100$).

Thermophysical properties of the HDPE-TEG system. To establish the effect of TEG on the thermophysical characteristics and the degree of crystallinity of the polymer matrix, the samples were investigated by the DSC method. Fig. 3 shows the results of DSC studies for composites based on HDPE in the temperature range from 380 to 440 K, since this is the most informative interval in which there is observed the polymer melting process. From the analysis of the data shown in Fig. 3 it can be seen that the amount of introduced TEG has a significant effect on the thermophysical characteristics of polymer filled systems. There is one endothermic maximum on the graph, which indicates that the HDPE is melting. For some composites, there is observed an additional maximum in the form of a shoulder, which is responsible for the melting of that part of the HDPE that is located at the polymer-graphite plate interface. Due to limited mobility upon contact with TEG, HDPE macromolecules cannot form a perfect crystal structure. Under conditions of space constraints (at the HDPE-TEG interface), there are formed defective crystallites, which require less energy to melt. Therefore, an additional endothermic maximum is observed at lower temperatures than the main HDPE maximum.

Table 1 shows the main thermophysical characteristics of the HDPE-TEG system.

Table 1 shows that TEGs affect the polymer matrix of HDPE. Thus, the melting point of composites decreases with increasing filler content. In the presence of TEG, smaller crystallites are formed, which require less energy to melt. In this case, the structure of the polymer matrix becomes more defective.



1 – 0 %; 2 – 0.2 %; 3 – 0.5 %; 4 – 0.8 %; 5 – 1 %

Figure 3 – Temperature dependence of heat capacity for polymer composites based on HDPE filled with TEG

Table 1 – Thermophysical characteristics of HDPE-based nanocomposites

Name	$T_m, ^\circ\text{C}$	$\Delta H_m, \text{J/h}$	$\chi_c, \%$
HDPE	415.0	217.9	75.4
HDPE +0.2 % TEG	413.5	211.2	73.1
HDPE +0.5 % TEG	412.1	203.7	70.5
HDPE +0.8 % TEG	410.7	196.2	67.9
HDPE +1.0 % TEG	408.5	186.6	64.6

The degree of crystallinity (χ_c) is one of the key characteristics of polymers and indicates the proportion of crystalline regions in the polymer. The degree of crystallinity of the systems under study can be calculated from the thermophysical data presented in Fig. 3 using formula (1) [10]:

$$\chi_c = \frac{\Delta H_m}{\Delta H_{m,c}} \cdot 100 \%, \quad (1)$$

where ΔH_m is the measured enthalpy of melting, $\Delta H_{m,c}$ is the enthalpy of melting of 100 % crystalline polymer (for HDPE, $\Delta H_{m,c}$ is 289 J/h [11]).

The values of the degree of crystallinity for the systems under study, calculated by formula (1), are given in Table. 1. This tendency to change the crystallinity of the matrix is well correlated with a decrease in the melting point of the system. It can be seen that the introduction of TEG into the polymer matrix leads to a significant decrease in the crystallinity of HDPE.

Thermal conductivity of the HDPE-TEG system.

Figure 4 shows the experimental results of the dependence of the thermal conductivity coefficient on the filler content for HDPE-TEG composites. Figure 4 illustrates that the concentration dependence of the thermal conductivity coefficient is nonlinear. A similar dependence of thermal conductivity was observed by the authors of [12] for polyethylene oxide – CNT

systems. The dependence in Fig. 4 can be conditionally divided into two areas: the first area (from 0 to 0.5 % TEG), where the thermal conductivity of the composite depends only on the structure of the polymer matrix; the second area (from 0.5 to 3 % TEG), where the increase in thermal conductivity is associated with the contribution of TEG to the thermal conductivity of the composite, which are characterized by a much higher coefficient of thermal conductivity compared to the polymer matrix.

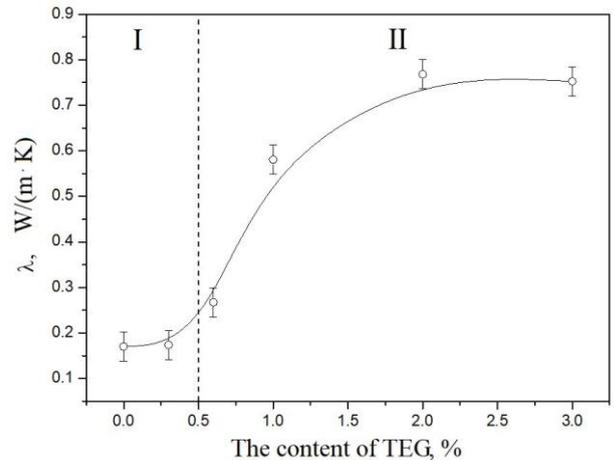


Figure 4 – Dependence of thermal conductivity on the content of TEG for systems based on HDPE

So, changes in thermal conductivity at low concentrations of fillers (Fig. 4) are associated with local compaction of the polymer matrix under the influence of uniformly distributed individual TEGs. The absence of the effect of increasing the thermal conductivity of the system at filler concentrations of 0.3–0.5 % is explained by phonon scattering at the filler-polymer interface [13] and by the fact that filler particles do not form a percolation cluster. In addition, the specified concentration range can be characterized by the presence of nanopores, which contributes to a decrease in the thermal conductivity of the composite due to a decrease in the thermal conductivity of the polymer and additional dissipation of energy on the inhomogeneities of the system [14].

The increase in thermal conductivity with a further increase in the filler content (Fig. 4) is associated with the contribution of TEGs, which are more thermally conductive composite component. Compared to a jump in electrical conductivity by several orders of magnitude at a TEG content of up to 1 %, the thermal conductivity of the HDPE-TEG system increases several times. Achievement of the thermal conductivity of the composite commensurate with the thermal conductivity of graphite plates is impeded by the presence of a developed polymer-filler interface, and the decisive factor is the energy loss due to the low thermal conductivity of the contacts [13]. A sharp increase in the thermal conductivity coefficient at a concentration of 0.6 % of TEG is associated with the percolation, in which the TEG plates form a heat-conducting network in the polymer matrix.

According to the theory of percolation, which considers the random distribution of the heat-conducting component in a non-conducting medium, the dependence of the thermal conductivity coefficient of the composite (λ_{ef}) on the TEG content can be described using the following equation [15]:

$$\lambda_{ef} \sim (p - p_c)^k, \text{ if } p > p_c, \quad (2)$$

where p is the content of the heat-conducting filler (TEG), p_c is the critical concentration of graphite plates (percolation threshold), k is the critical index of thermal conductivity.

Applying the least squares method and equation (2) to describe the experimental data on the concentration dependence of thermal conductivity (Fig. 4), we determined the percolation threshold p_c and the critical index k , which characterizes the structural organization of the nanofiller in the composite and depends on its size and shape. The value of the percolation threshold (p_c) for the HDPE-TEG system is 0.6 %, and $k = 0.28 \pm 0.03$. The percolation threshold was found to be lower than for most polymer-carbon filler systems. Thus, for the ethylene terpolymer-CNT system, p_c was 2.2 % [16], and for the PE-CNT system, the percolation threshold was 0.72 % [9]. Such discrepancies are explained by different methods of preparation of polymer composites, sizes and types of carbon fillers and indicate different processes of their aggregation.

Mechanical properties of HDPE-TEG systems.

In addition to the effect on the microstructure and thermophysical properties, modification of the HDPE matrix using thermally expanded graphite plates leads to a significant improvement in the mechanical characteristics of the materials obtained.

To study the effect of the filler on the mechanical characteristics of the HDPE-TEG system, there was researched the tensile strength of the obtained materials. Figure 5 demonstrates the dependence of the tensile strength (σ_{it}) on the content of thermally expanded graphite. From the obtained dependence it can be seen that with an increase in the filler content, the tensile strength of the system increases. At the same time, with a content of 5 % in the material, the tensile strength increases by more than 40 %, this makes this material promising for manufacturing structural materials for transportation systems.

Figure 5 shows that with an increase in the filler content, the tensile strength of the studied systems increases from 30.5 MPa for unfilled HDPE to 42.8 for the HDPE – 5 % of TEG system. The increase in mechanical characteristics in the obtained materials can be explained by the contribution from the filler, which has a higher strength than the polymer matrix. Fig. 5 also shows that the increase in mechanical tensile strength is nonlinear. In the range of filler concentrations of 0.5–1.0 %, there is observed an abrupt increase in strength. This is due to the formation of a reinforcing mesh inside the material with filler particles,

which, interacting with each other and the matrix, significantly enhance its mechanical characteristics [17].

The prospect of using polymer composites to create structural materials for transportation systems for various purposes is associated with a set of their characteristics, such as the necessary thermophysical and mechanical properties, high corrosion resistance, relatively low specific gravity, etc. [18].

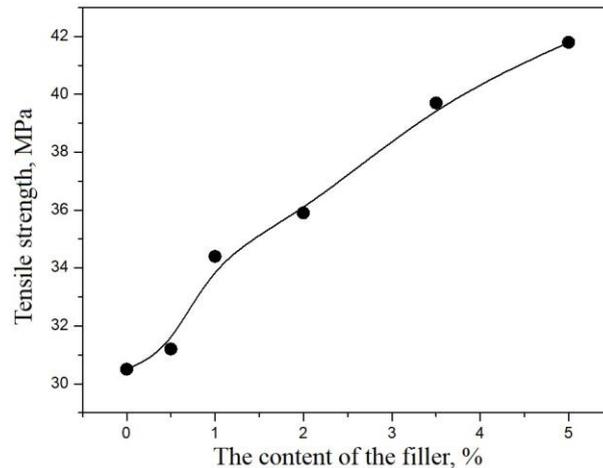


Figure 5 – Dependence of thermal conductivity on the TEG content for HDPE based systems

Characterizing the developed materials in general, it should also be noted that they have high corrosion resistance. This is very important when creating structural materials that are operated in conditions of exposure to aggressive media. Traditionally, in such situations, metals are used as a material. A comparative assessment illustrates that their cost is tens times higher than the cost of the developed polymer composites.

Conclusions

As a result of the research performed, there has been investigated the effect of TEG on the microstructure and functional properties of polymer composites based on high-density polyethylene. It has been shown that the introduction of TEG leads to a decrease in the degree of crystallinity and the melting temperature of systems based on HDPE, and it is a consequence of the destruction or an increase in the defectiveness of the crystal structure of the polymer matrix under the influence of TEG. It has been found that the use of carbon fillers for polymer matrices leads to an extreme effect on the properties in composites at their ultra-low concentrations (~ 0.5–1.0 %). An extreme change in the properties of polymer composites is a consequence of the TEG formation in the polymer matrix of a percolation cluster, that is, a network with a filler that permeates the entire volume of the material. This process can be described in terms of percolation theory. The survey of conductivity has revealed the percolation threshold of thermal conductivity for these HDPE-TRG systems, which is 0.6 %. According to the results of microscopic studies, there has been confirmed the value of the percolation threshold obtained from the results of thermal conductivity. It is shown that at a

content of 0.6 % of TEG, there is formed a continuous cluster. There is recorded a nonlinear increase in the mechanical strength in the studied system, and this is a consequence of the formation of a reinforcing mesh with a filler inside the polymer matrix. There has been revealed high mechanical strength, which increases by more than 40 % at 5 % filler content.

Thus, the application of the mixing method using a piston extruder made it possible to obtain polymer composite materials, which, according to their characteristics, are promising for use in transportation systems. A prospect for the further development of this direction is the use of a combined type of filler, for example, a mixture of thermally expanded graphite and carbon nanotubes, to make polymer composites. The synergistic effect of carbon fillers of various shape rheometry will enable to obtain the necessary functional characteristics of the obtained materials at significantly lower concentrations.

The article presents the results of studies carried out within the framework of the state budget research theme "Creation of new multifunctional polymer nanocomposites containing carbon nanotubes" (State Registration Number 0121U100658), funded by the Ministry of Education and Science of Ukraine.

References

- [1] Hsissou, R et.al. 2021, 'Polymer composite materials: A comprehensive review', *Composite Structures*, vol. 262, Article 113640.
- [2] Kumar, VV et.al. 2019, 'A Review of Recent Advances in Nanoengineered Polymer Composites', *Polymers*, 11(4):644.
- [3] Gantayat, S et.al. 2015, 'Expanded graphite as a filler for epoxy matrix composites to improve their thermal, mechanical and electrical properties', *New Carbon Materials*, 30(5): 432–437. doi: 10.1016/S1872-5805(15)60200-1
- [4] Tang, YJ, Lin, YX, Jia, YT & Fang, GY 2017, 'Improved thermal properties of stearyl alcohol/high density polyethylene/expanded graphite composite phase change materials for building thermal energy storage', *Energy and Buildings*, 153: 41–49.
- [5] Mochane, MJ & Luyt, AS 2015, 'The effect of expanded graphite on the flammability and thermal conductivity properties of phase change material based on PP/wax blends', *Polymer Bulletin*, vol. 72, pp. 2263–2283.
- [6] Tavman, I et.al. 2011, 'Measurement of heat capacity and thermal conductivity of HDPE/expanded graphite nanocomposites by differential scanning calorimetry', *Archives of Materials Science and Engineering*, vol. 50, no. 1. pp. 56–60.
- [7] Sobolciak, P et.al. 2020, 'Thermally Conductive Polyethylene/Expanded Graphite Composites as Heat Transfer Surface: Mechanical, Thermo-Physical and Surface Behavior', *Polymers*, vol. 12, no. 12, p. 2863.
- [8] Sementsov, YuI, Revo, SL & Ivanenko, KO 2016, 'Thermoexpanded graphite', the acad. of the NAS of Ukraine M.T. Kartel, Chuiko Institute of Surface Chemistry of NAS of Ukraine; Taras Shevchenko National University of Kyiv, p. 241. [in Ukrainian]
- [9] Dinzhos, RV, Fialko, NM & Lysenkov, EA 2014, 'Analysis of the Thermal Conductivity of Polymer Nanocomposites Filled with Carbon Nanotubes and Carbon Black', *J. of Nano-Electron. Phys.*, vol. 6, no. 1, p. 01015.
- [10] Bershtein, VA & Egorov, VM 1990, *Differential scanning calorimetry in the physicochemistry of polymers*, Chemistry, Leningrad, 256 p. [in Russian]
- [11] Mirabella, FM & Bafna, A 2002, 'Determination of the crystallinity of polyethylene/ α -olefin copolymers by thermal analysis: Relationship of the heat of fusion of 100 % polyethylene crystal and the density', *Polymer Physics*, vol. 40, no. 15. pp. 1637–1643.
- [12] Lysenkov, EA & Klepko, VV 2015, 'Characteristic Features of the Thermophysical Properties of a System Based on Polyethylene Oxide and Carbon Nanotubes', *Journal of Engineering Physics and Thermophysics*, vol. 88, pp. 1008–1014.
- [13] Xu, Y, Ray, G & Abdel-Magid, B 2006, 'Thermal behavior of single-walled carbon nanotube polymer matrix composites', *Composites Part A: Applied Science and Manufacturing*, vol. 37, no. 1, pp. 114–121.
- [14] Garkusha, OM etc. 2011, 'Structural features and properties of polymeric nanocomposites at low concentrations of filler', *Chemistry, physics and surface technology*, vol. 1, no. 1, pp. 103–110. [in Ukrainian]
- [15] Stauffer, D & Aharony, A 1994, *Introduction to percolation theory*, Taylor and Francis, London, 272 p.
- [16] Kim, B-W, Park, S-H, Kapadia, RS & Bandaru, PR 2013, 'Evidence of percolation related power law behavior in the thermal conductivity of nanotube/polymer composites', *Applied Physics Letters*, 102, 243105; doi: 10.1063/1.4811497. View online: <http://dx.doi.org/10.1063/1.4811497>
- [17] Hadzreel, MRAM & Aisha, ISR 2013, 'Effect of Reinforcement Alignment on the Properties of Polymer Matrix Composite', *Journal of Mechanical Engineering and Sciences*, vol. 4, pp. 548–554.
- [18] Wang, Q et. al. 2012, 'A review on application of carbonaceous materials and carbon matrix composites for heat exchangers and heat sinks', *International Journal of Refrigeration*, vol. 38, pp. 7–26.

УДК 538.9: 536.21: 536.6

Мікроструктура та властивості полімерних композитних матеріалів на основі поліетилену та терморозширеного графіту для систем транспортування

Е.А. Лисенков, Л. П. Хименко

*Чорноморський національний університет ім. Петра Могили;
10, вул. 68 Десантників, м. Миколаїв, 54003, Україна*

Методами оптичної мікроскопії, диференціально сканувальної калориметрії та механічного аналізу вивчено вплив терморозширеного графіту (ТРГ) на мікроструктуру та функціональні властивості полімерних композитів на основі поліетилену високої густини (ПЕВГ). Доведено ефективність методу змішування полімерних композитів за допомогою поршневого екструдера, який забезпечує рівномірніший розподіл наповнювача у полімерній матриці. Показано, що введення ТРГ приводить до зниження ступеня кристалічності та температури плавлення систем на основі поліетилену високої густини, що є наслідком руйнування або зростання дефектності кристалічної структури полімерної матриці під впливом ТРГ. При введенні 1 % ТРГ температура плавлення знизилася з 415.0 до 408.5 К. При введенні 3 % ТРГ теплопровідність зросла з 0.18 Вт/(м·К) (для ПЕВГ) до 0.76 Вт/(м·К). Екстремальна зміна теплопровідності полімерних композитів є наслідком утворення ТРГ у полімерній матриці перколяційного кластеру, тобто сітки із наповнювача, яка пронизує весь об'єм матеріалу. У результаті проведених досліджень провідності було визначено поріг перколяції теплопровідності для даних систем ПЕВГ–ТРГ, який становить 0.6 %. Мікроскопічні дослідження підтвердили утворення перколяційного кластеру, отриманого за результатами теплопровідності. Показано, що при вмісті 0.6 % ТРГ, утворюється неперервний кластер. Утворення цього кластеру підтверджується і механічними дослідженнями. Зафіксовано зростання механічної міцності, яка при 5 % вмісті наповнювача збільшується з 30.5 МПа (для ПЕВГ) до 42.8 МПа, що є перспективним для застосуванні даних матеріалів у системах транспортування.

Ключові слова: *перколяція, поліетилен високої густини, полімерний композитний матеріал, терморозширнений графіт.*